

**NUCLEAR FUEL BODY INCLUDING TUNGSTEN NETWORK**  
**AND METHOD OF MANUFACTURING THE SAME**

**FIELD OF THE INVENTION**

5           The present invention relates to a nuclear fuel  
body comprising tungsten network and a method for  
manufacturing the same, more particularly, a nuclear  
fuel body in which tungsten network is continuously  
formed over the entire or some parts of a sintered body  
10 and a method for manufacturing the same.

**BACKGROUND**

A nuclear power plant utilizes the fission energy  
produced in nuclear fuel. Tens of - hundreds of fuel  
15 bodies composed of nuclear fuel materials are loaded in  
a zirconium-based tube, which is then seal-welded to  
fabricate a fuel rod. Tens of - hundreds of fuel rods  
are assembled to make a bundle and these bundles are  
loaded in water reactors. Heat generated in the fuel  
20 body is transferred through the zirconium-based tube to  
coolant flowing around the fuel rods.

For a nuclear fuel, cylindrical or spherical  
shape fuel body fabricated by sintering and processing



the individual oxide or the mixture of oxides of uranium(U), plutonium(Pu), thorium(Th), etc are used. Uranium dioxide( $\text{UO}_2$ ) is generally used to fabricate a fuel body and one or more materials selected from a group consisting of oxides of Pu, Th, Gd, etc can be added to  $\text{UO}_2$ . Particularly,  $(\text{U}, \text{Pu})\text{O}_2$ ,  $(\text{U}, \text{Th})\text{O}_2$ ,  $(\text{U}, \text{Gd})\text{O}_2$ ,  $(\text{U}, \text{Pu}, \text{Gd})\text{O}_2$  or  $(\text{U}, \text{Th}, \text{Pu})\text{O}_2$  is preferably used.

The most preferably used nuclear fuel is the uranium oxide sintered body, which is prepared by the steps of mixing lubricant with uranium oxide powder, preparing slug by preliminary processing with 1 ton/ $\text{cm}^2$  pressure, preparing granules by crushing the slug, pressing the granules into green pellet having about 50% TD (theoretical density), and sintering the green body at 1600-1800°C for 2-4 hours in hydrogen-containing gas. The produced uranium oxide sintered body has a cylindrical shape and 95% density based on theoretical density. The microstructure of the sintered body is represented in FIG. 1. As shown in FIG. 1, the structure of the sintered body is constructed by polygonal grains having 3~20  $\mu\text{m}$  in diameter.

In like manner,  $(\text{U}, \text{Pu})\text{O}_2$  or  $(\text{U}, \text{Th})\text{O}_2$  sintered body is prepared by mixing plutonium oxide powder or thorium oxide powder with uranium oxide powder and then by following the above steps as used for preparing the



uranium oxide sintered body.  $(U,Gd)O_2$  sintered body is also prepared by mixing gadolinium oxide powder with uranium oxide powder and then by following the steps as used for preparing the uranium oxide sintered body. In order to grow grains of nuclear fuel sintered body, one or more oxides selected from a group consisting of Nb, Ti, Si, Mg and Al oxides are added to uranium oxide.

$UO_2$  has been generally used as a nuclear fuel material since it has a high melting point and reacts negligibly with coolant(water). But,  $UO_2$  has comparatively low thermal conductivity ( $2\sim 5$  W/m K). If a nuclear fuel material has low thermal conductivity, heat generated by nuclear fission cannot be transferred to coolant right away, resulting in an increase in the temperature of fuel body. The temperature of fuel body is highest at the center and lowest at the surface. The difference in temperature between the surface and the center of the sintered body is inversely proportional to thermal conductivity. Therefore, the center temperature of fuel body increases as thermal conductivity decreases. The center temperature of fuel body in normally burning nuclear fuel rods ranges from  $1000^{\circ}C$  to  $1500^{\circ}C$ , but if it is not in a normal operation, the temperature can go over  $2800^{\circ}C$ , which is the melting point of  $UO_2$ .



Since the nuclear fuel sintered body keeps high temperature, all the temperature-depending reactions are accelerated, resulting in the lowering of material performance. The higher the burning temperature is,  
5 the lower the performance becomes. In addition, a high temperature in sintered body lowers the safety margin in anticipated nuclear power plant accidents. In case of rapid elevation of the generating power of fuel body, the center temperature might increase up to over the  
10 melting point of  $\text{UO}_2$ . In order to prevent this problem, the generating power ought to be limited, with suffering an economic loss.

The thermal conductivities of  $(\text{U,Pu})\text{O}_2$ ,  $(\text{U,Gd})\text{O}_2$  or  $(\text{U,Th})\text{O}_2$  are as good as or worse than that of  $\text{UO}_2$ .  
15 Especially, the thermal conductivity of  $(\text{U,Gd})\text{O}_2$  is much worse than that of  $\text{UO}_2$ , suggesting that the temperature of  $(\text{U,Gd})\text{O}_2$  sintered body is much higher than that of  $\text{UO}_2$  sintered body under the condition of having same generating power. To solve the problem,  
20 the generating power of  $(\text{U,Gd})\text{O}_2$  sintered body ought to be more limited than that of  $\text{UO}_2$  fuel body.

Thus, the present invention prepared a novel nuclear fuel sintered body in which tungsten network  
25 having a high melting point and excellent thermal conductivity is continuously formed in order to make up



for such disadvantage that the nuclear fuel sintered body has a low thermal conductivity.

#### SUMMARY OF THE INVENTION

5           It is an object of this invention to provide a nuclear fuel body containing tungsten network having enhanced thermal conductivity and a preparation method thereof.

#### 10           BRIEF DESCRIPTION OF THE DRAWINGS

The application of the preferred embodiments of the present invention is best understood with reference to the accompanying drawings, wherein:

15           FIG. 1 is an optical microphotograph showing the microstructure of the uranium oxide sintered body provided by the prior art;

20           FIG. 2 is an optical microphotograph showing the microstructure of the uranium oxide sintered body comprising tungsten network of the present invention;

FIG. 3 is an optical microphotograph showing the microstructure of the preliminary sintered body in



which tungsten particles are uniformly dispersed;

FIG. 4 is a graph showing the oxygen partial pressure for tungsten oxidation;

5

FIG. 5 is an optical microphotograph showing the microstructure of the network of liquid tungsten oxide obtained by heat-treatment of the preliminary sintered body in the presence of an oxidizing gas.

10

#### DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

The present invention provides a nuclear fuel body containing tungsten network and a method for manufacturing the same.

15

Further features of the present invention will appear hereinafter.

The present invention provides a nuclear fuel body comprising fuel grains and tungsten network. Particularly, the nuclear fuel body comprises tungsten network that is continuously formed between fuel grains and thus envelops fuel grains (see FIG. 2).

20

As shown in FIG. 2, tungsten network is continuous between fuel grains throughout the entire uranium oxide sintered body and acts as a heat-

25



conducting channel. The lines - metal network - shown in two-dimensional photograph mean the planes in three-dimensions, and the polygons - fuel grains - in two-dimensional photograph are the polyhedrons in three-dimensions. Thus, the metal tungsten has a shape which envelopes the polyhedral grains. As tungsten network is completely formed, nuclear fuel materials are in polyhedral tungsten and such tungsten polyhedrons are continuously gathered together to constitute a sintered body. The sizes of the tungsten polyhedrons are corresponding to those of fuel grains.

The fuel grain size depends on the conditions of its preparation method. The fuel grains become larger as additives are added to nuclear fuel materials. As an additive, Nb, Ti, Al, Si or Mg oxide can be used. The grain size of nuclear fuel sintered body provided by the present invention is between 5 and 500  $\mu\text{m}$  in diameter in two-dimensional photograph. Thus, the size of one unit of tungsten network is also between 5 and 500  $\mu\text{m}$  in diameter and the tungsten channel between two neighboring fuel grains has thickness ranging from 0.1 to 20  $\mu\text{m}$ .

The amount of tungsten in the fuel body increases with increasing the thickness of tungsten channel and with decreasing the size of fuel grain. On the contrary, the amount of nuclear fuel material in



sintered body decreases with increasing the amount of tungsten therein and so does the fission energy produced in fuel body, causing a drop in economical efficiency. Therefore, it is desirable to enhance the thermal conductivity of fuel body by a small amount of tungsten. The nuclear fuel sintered body provided by this invention has the amount of tungsten ranging from 0.2 to 50% by weight of fuel body.

The melting point of tungsten is  $3400^{\circ}\text{C}$  and the melting point of uranium oxide is  $2800^{\circ}\text{C}$ . That is the reason why tungsten is chosen as a network material. Tungsten alloys containing other metals up to about 10% by weight can be used as network materials unless they do not significantly decrease the melting point of tungsten alloys.

The nuclear fuel body of the present invention is characterized by being composed of nuclear fuel materials and tungsten network. Uranium oxide or uranium oxide mixture, prepared by mixing one selected from a group consisting of gadolinium oxide, plutonium oxide and thorium oxide with uranium oxide, can be used for the nuclear fuel material. If tungsten is dispersed in a form of isolated particles with no tungsten network, tungsten cannot act as a heat-conducting channel. The nuclear fuel body of the present invention is characterized by having tungsten



network, not by tungsten dispersed in a form of isolated particles.

There are two steps of transferring the heat generated from nuclear fuel materials in the nuclear fuel body of the present invention. The heat generated in the nuclear fuel grains by nuclear fission of uranium is conducted to the neighboring tungsten network through uranium oxide (step 1), and the heat is further conducted through the tungsten network having relatively high thermal conductivity and being continuous throughout the entire sintered body, and thereby the heat generated in the center of the body can be conducted to the surface of the body (step 2). The thermal conductivity of tungsten is higher by 25 times than that of uranium oxide. Thus, the nuclear fuel body of the present invention has enhanced thermal conductivity.

Tungsten network channel is not a line but a plane in three-dimensions, so point damage of a plane does not much degrade the heat conduction through the tungsten channel.

The nuclear fuel body of the present invention includes the tungsten network throughout the entire fuel body. Some variations are possible in the spirit



of this invention. The tungsten network can be formed in local regions of the fuel body. Especially, it is possible to prepare a cylindrical fuel body in which an inner cylinder has tungsten network but the outside  
5 ring does not. Conversely, it is also possible to prepare a fuel body in which tungsten network is formed only in the outside ring.

In order to supplement the low thermal conductivity of conventional nuclear fuel bodies, every  
10 possible factor that might decrease the thermal conductivity have been restricted. Since the nuclear fuel body of the present invention comprises tungsten network, which means heat is conducted through the tungsten network, thermal conductivity does not much  
15 affected by the change in the oxygen/uranium ratio and the density of the fuel body. That is, the nuclear fuel body of the present invention can be used in the wide range of oxygen/uranium ratio and density.

20 The present invention also provides a method for manufacturing a nuclear fuel sintered body containing tungsten network.

Particularly, the present invention provides a preparation method for a nuclear fuel body comprising  
25 the following steps:

Preparing a green body composed of nuclear fuel



powder and tungsten-containing powder (Step 1);

Preparing a preliminary sintered body, in which tungsten particles are dispersed, by heating the above green body in a reducing gas (Step 2);

5       Forming liquid tungsten oxide network in the preliminary sintered body by heating the preliminary sintered body in an oxidizing gas (Step 3); and

Preparing a nuclear fuel body comprising solid tungsten network by heating in a reducing gas the above  
10 preliminary sintered body having liquid tungsten oxide network (Step 4).

The details of the above preparation method are described in the followings.

15       In the above step 1, a powder mixture is prepared by mixing uniformly fuel powder and tungsten-containing powder. The tungsten-containing powder is one selected from the group consisting of tungsten, tungsten oxide and mixtures thereof. The amount of tungsten-  
20 containing powder is between 0.2 and 50% by weight of the powder mixture. And then, a green body is prepared by loading and pressing the powder mixture in a mold. For the fuel powder above, uranium oxide or the mixture prepared by mixing uranium oxide and one or more  
25 selected from a group consisting of gadolinium oxide, plutonium oxide and thorium oxide can be used. The



tungsten-containing powder can include other metal elements less than 10% by weight of tungsten.

5 In step 2, a preliminary sintered body is prepared by heating the green body at 1,100~2,000°C in a reducing gas. As a reducing gas that is able to reduce tungsten oxide to tungsten, hydrogen gas or gas mixture prepared by mixing hydrogen gas and one or more selected from a group consisting of nitrogen, inert gas, carbon dioxide, carbon monoxide and steam can be used.

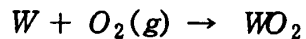
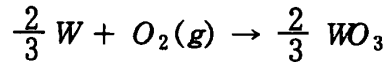
10 When uranium oxide is used as fuel powder, the preliminary sintered body has such microstructure that metal tungsten is uniformly dispersed in a form of isolated particles in  $\text{UO}_2$  matrix (see FIG. 3). The preliminary sintered body, thus, does not have a good thermal conductivity since tungsten that is dispersed  
15 in a form of particles cannot act as a heat-conducting channel.

In step 3, liquid network of tungsten oxide is formed in the fuel body by heating the preliminary  
20 sintered body in an oxidizing gas that is able to oxidize tungsten to tungsten oxide ( $\text{WO}_3$ ). At this time, the heating temperature is 1,100~1,800°C and the oxidizing gas ought to be a gas which has oxygen partial pressure controlled at least to oxidize  
25 tungsten to tungsten oxide. The oxygen partial pressure of the oxidizing gas is calculated from the



free energy of tungsten oxidation reaction. Tungsten oxide includes  $WO_3$  and  $WO_2$  and the oxidation reaction is expressed by the following <Reaction Formula 1>.

5 <Reaction Formula 1>



The relation between the Gibbs free energy and oxygen partial pressure is expressed by the following <Reaction Formula 2>.

10

<Reaction Formula 2>

$$\Delta G^0(T) = -RT \ln K = RT \ln P_{O_2}$$

15 In the above reaction formula,  $\Delta G^0(T)$  is the Gibbs free energy,  $R$  is the gas constant,  $T$  is the absolute temperature,  $K$  is the reaction constant, and  $P_{O_2}$  is the equilibrium oxygen partial pressure.

20 The Gibbs free energy ( $\Delta G^0(T)$ ) is well known as a function of temperature, so the  $RT \ln P_{O_2}$  can be calculated based on that. Equilibrium oxygen partial pressure is calculated from  $RT \ln P_{O_2}$ , which is represented in FIG. 4 as a function of temperature.



When the oxidizing gas having higher oxygen partial pressure than the equilibrium oxygen partial pressure between W and  $\text{WO}_3$  is used at the heating process, tungsten is oxidized to tungsten oxide ( $\text{WO}_3$ ). The  
5 melting point of  $\text{WO}_3$  is about  $1480^\circ\text{C}$ . So, when an oxidizing gas has higher oxygen partial pressure than required for forming  $\text{WO}_3$  and the heating temperature is over  $1480^\circ\text{C}$ , the liquid tungsten oxide can be obtained. Liquid tungsten oxide penetrates into the grain  
10 boundary of uranium oxide and thus forms network of liquid  $\text{WO}_3$ , which is confirmed by investigating the microstructure of preliminary sintered body (see FIG. 5). As shown in FIG. 5,  $\text{WO}_3$  is formed between polygonal grains, suggesting that network of liquid  
15 tungsten oxide is formed in the preliminary sintered body.

On the other hand, it is also observed that tungsten oxide reacts with uranium oxide to form (uranium + tungsten) oxide. This (uranium + tungsten)  
20 oxide melts even under  $1480^\circ\text{C}$  and then penetrates into the grain boundary of fuel to form network of liquid tungsten oxide, so the microstructure of the sintered body is similar to that shown in FIG. 5.

The heating temperature for the preliminary  
25 sintered body is preferably over  $1100^\circ\text{C}$  and the oxygen partial pressure of an oxidizing gas is required to be



higher than the equilibrium oxygen partial pressure of W and  $WO_3$  reaction.

In order to regulate the oxygen partial pressure of an oxidizing gas, carbon dioxide or steam is used, or one or more methods selected from a group consisting of regulating the mixing ratio of carbon dioxide to carbon monoxide, mixing ratio of hydrogen to steam, mixing ratio of hydrogen to carbon dioxide, mixing ratio of inert gas or nitrogen to oxygen, etc., can be further used.

At last, in step 4, a nuclear fuel body comprising solid tungsten network is prepared by heating the above preliminary sintered body having liquid tungsten oxide network in a reducing gas. At this time, the heating temperature is  $1,100\sim 2,000^{\circ}\text{C}$ . The liquid tungsten oxide reduces to solid tungsten by heating in a reducing gas the preliminary sintered body having liquid tungsten oxide network, which results in the preparation of a nuclear fuel body comprising solid tungsten network. It is preferable that the oxygen partial pressure of the reducing gas is lower than the equilibrium oxygen partial pressure of W and  $WO_2$  reaction shown in FIG. 4. The reducing gas is hydrogen and further comprises one selected from a group consisting of nitrogen, inert gas, carbon dioxide, carbon monoxide, steam and mixtures thereof.



The above preparation method provides a fuel body comprising the tungsten network throughout the entire fuel body. The green body that consists of the uniform powder mixture of fuel and tungsten-containing powder is processed according to the steps 2 to 4. However, an inhomogeneous green body can be processed according to the steps 2 to 4, because liquid tungsten oxide can penetrate into a long distance over the entire body if a sufficient time is provided for the heating in the step 3. Thus, even a green body in which tungsten-containing powder is extremely segregated can also lead to the same fuel body comprising tungsten network according to the same procedures of the above steps 2 to 4. Two cases are described where the tungsten-containing powder is located in the center and at the surface of the green body.

Firstly, in step 1 a green body in which tungsten-containing powder is located in the center and fuel powder is located in the surroundings is prepared. And then, a nuclear fuel body is prepared using the green body according to the same procedures of the above steps 2-4. In step 2, a preliminary sintered body contains tungsten particles in the center. In step 3, tungsten is oxidized to tungsten oxide and liquid tungsten oxide penetrates into grain boundaries



of the surrounding uranium oxide, forming eventually a network of liquid tungsten oxide over the entire body. In step 4, the network of liquid tungsten oxide is reduced to tungsten network. As a result, a sintered  
5 body in which tungsten network is continuously formed through the entire body is prepared.

Secondly, in step 1 a green body is prepared by pressing the fuel powder only, and then the tungsten-  
10 containing powder, confined by a cap, is put on the surface of the green body. In step 2, a preliminary sintered body where tungsten particles adhere to its surface is prepared by heating the green body in a reducing gas. After then, a nuclear fuel body is  
15 prepared using the preliminary sintered body according to the same procedures of the above steps 3-4. The preliminary sintered body prepared in the above step 2 has tungsten particles adhered to the surface, which are oxidized in step 3. Liquid tungsten oxide on the  
20 surface penetrates into grain boundaries of the preliminary sintered body to form liquid network continuously through the entire body. At last, in step 4, the liquid network of tungsten oxide reduces to solid tungsten network. As a result, a nuclear fuel  
25 body containing tungsten network that is formed throughout the entire fuel body is prepared.



The method for manufacturing a nuclear fuel body of the present invention can provide a possible way to form tungsten network not only throughout the entire body but also in local regions. For example, it is possible to prepare a cylindrical fuel body in which an inner cylinder has tungsten network but the outside ring does not. Conversely, it is also possible to prepare a fuel body in which tungsten network is formed only in the outside ring. Particularly, to manufacture the fuel body having the tungsten network only in the inner cylinder, the powder mixture of uranium oxide and tungsten-containing powder is loaded in the inner cylinder void of a mold and uranium oxide powder is loaded in the outside ring void, and thereafter powders are pressed in a mold. Then a preliminary sintered body is prepared by sintering the said green body in a reducing gas. Tungsten particles are present in uranium oxide matrix of the inner cylinder part of the preliminary sintered body but not in outside ring. By heating the preliminary sintered body in an oxidizing gas in accordance with the preparation method of the present invention, tungsten network is formed in inner cylinder of the sintered body, but not in outside ring. When the positions of both powders are changed each other, tungsten network is formed in outside ring, but



not in inner cylinder.

#### EXAMPLES

Practical and presently preferred embodiments of  
5 the present invention are illustrative as shown in the  
following Examples.

However, it will be appreciated that those skilled  
in the art, on consideration of this disclosure, may  
make modifications and improvements within the spirit  
10 and scope of the present invention.

##### Example 1: Preparation of a nuclear fuel sintered body

Tungsten powder was added to uranium oxide powder  
in the quantity of 9% by weight, and then two powders  
15 were mixed. The powder mixture was loaded in a  
cylindrical mold, and then pressed under 3 t/cm<sup>2</sup> to make  
a green body. The green body was sintered at 1700°C  
for 4 hours in hydrogen gas to make a preliminary  
sintered body. The preliminary sintered body was  
20 heated at 1480°C for 1.5 hours in carbon dioxide gas.  
Finally, the above preliminary sintered body was  
reduced at 1650°C in hydrogen gas for 2 hours to  
produce the nuclear fuel body.



Experimental Example 1: Investigation on microstructure  
of the nuclear fuel body

The microstructure of the nuclear fuel body produced in the above Example 1 was investigated by an  
5 optical microscope (200-500X magnification). The results were shown in FIG. 2, 3 and 5.

FIG. 2 shows the microstructure of the nuclear fuel body comprising tungsten network produced by heating the preliminary sintered body containing liquid  
10 tungsten oxide network in a reducing gas. It is confirmed that tungsten network is continuously formed in the entire sintered body and the tungsten network acts as a channel for heat conduction.

FIG. 3 shows the microstructure of the preliminary sintered body in which tungsten particles  
15 are dispersed uniformly. The tungsten particles are not connected but scattered, so they do not act as a channel for heat conduction, suggesting that the body has low thermal conductivity.

FIG. 5 shows the microstructure of the preliminary sintered body comprising liquid tungsten oxide network and uranium oxide grains. Heating the preliminary sintered body in an oxidizing gas produces  
20 this microstructure. It is confirmed that  $WO_3$  is formed  
25 between polygonal grains and liquid tungsten oxide



network is formed in entire sintered body.

Experimental Example 2: Measurement of the thermal diffusivity of the nuclear fuel body

- 5           The thermal diffusivity of the nuclear fuel body prepared in the above Example 1 was measured by the laser flash method, and the thermal diffusivity of the pure uranium oxide body was also measured for comparison. The results are shown in Table 1.

10   <Table 1>

Thermal diffusivity of nuclear fuel body comprising tungsten network

Temperature (°C)	Thermal diffusivity ( $\times 10^{-6} \text{ m}^2/\text{sec}$ )		Ratio of A to B (A/B)
	Nuclear fuel body comprising tungsten network (A)	Pure uranium oxide body (B)	
22	5.271	3.617	1.46
200	4.450	2.772	1.61
400	3.282	1.945	1.69
600	2.629	1.487	1.77
800	2.239	1.238	1.81
1000	1.978	1.085	1.82
1200	1.778	0.956	1.86
1400	1.619	0.879	1.84



As shown in Table 1, the thermal diffusivity of the nuclear fuel body comprising tungsten network of the present invention was higher by 70-80% than that of the pure uranium oxide body. Particularly, the enhanced ratio (A/B) of thermal diffusivity was 1.46-1.86 at 22-1200°C.

#### INDUSTRIAL APPLICABILITY

As explained hereinbefore, the nuclear fuel body in which tungsten network is continuously formed between grains throughout the entire body or in some specific regions can contribute to enhance the safety of a nuclear reactor by enhancing the thermal conductivity of the fuel body and decreasing the temperature of burning fuel body.